Towards gas sensing without spectroscopy using mid-infrared optical parametric oscillators

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Abstract: We introduce a method for gas sensing without performing direct spectrum measurement using broadband mid-infrared optical parametric oscillators, and experimentally demonstrate proof-of-concept carbon dioxide sensing.

Mid-infrared (IR) is the principal spectral region of interest for molecular sensing where most molecules exhibit strong spectral fingerprints. Myriad applications such as medical breath analysis [1] and environmental sensing [2] have motivated extensive efforts on developing broadband coherent spectrometers in the mid-IR for multi-species gas sensing, for instance using dual-comb spectroscopy [3–5]. However, achieving a low-noise, coherent broadband spectrometer in the mid-IR typically requires addressing substantial challenges regarding the source [6] and spectroscopy technique [7]. Mid-IR photodetectors have also imposed barriers (sensitivity, response time, etc.) in making such systems available for many applications. On the other hand, broadband optical parametric oscillators (OPOs) offer a unique opportunity for multi-species molecular sensing. While broadband OPOs have been one of the most promising sources of broadband mid-IR frequency combs [8], their operations can be extremely sensitive to spectral features of intracavity losses and dispersion [9]. This feature can be utilized as a sensitive tool for multi-species molecular sensing without requiring a spectrometer or even a photodetector in the mid-IR.

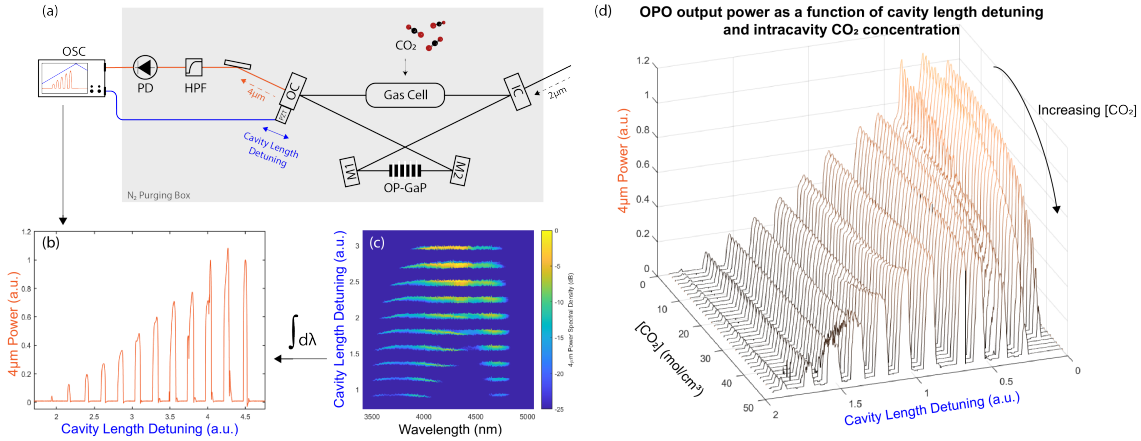

Fig. 1. (a) Schematic of the experimental setup. (b) Example output signal from the photodetector as a function of cavity length. The peaks correspond to various resonant conditions in doubly-resonant OPOs. (c) OPO output power spectral density at various cavity length detuning values. (d) Output signal (at 4 μm) peaks evolution as a function of CO_2 concentration.

In this work, we introduce and experimentally demonstrate an alternative method for broadband gas sensing based on the dependence of the OPO power on the intracavity gas concentrations without analyzing the spectrum and show that this information can be extracted with a near-IR photodetector. The schematic of the experimental setup is shown in Fig. 1(a). We utilize a femtosecond optical parametric oscillator (OPO) with an output centered around 4 μm [10], with a typical output power as a function of cavity detuning shown in Fig. 1(b), and output spectrum as a function of cavity detuning in Fig. 1(c). Sensing is performed using the gas sample in the cavity of this OPO, which is contained in a box to flush out atmospheric gases with nitrogen. We placed a 10-cm pathlength glass gas cell with silicon and calcium fluoride windows within the 4-μm OPO cavity path after the OP-GaP crystal. Given that the output power and the spectrum of the OPO are cavity-length dependent with extreme sensitivity [9, 11], a wealth of information about the spectral features of gas molecules inside the gas cell can be extracted by monitoring the output power as a function of cavity roundtrip detuning (Fig. 1b) with a single photodetector (Thorlabs PDAVJ5). Each point on the "detuning peaks" is an integral of the OPO power spectrum

across all wavelengths of the output spectra at various cavity roundtrip lengths, as shown in Fig.1(c). When the target molecule absorption lines overlap with the OPO spectrum, spectral information is reflected in the shapes of these peaks due to absorption and dispersion. These features can be very sensitive to the magnitude of absorption and dispersion and their spectral distribution, which we utilize as the basis for molecular sensing.

We perform proof-of-concept measurements of carbon dioxide demonstrating changes in output signal detuning peaks as a function of gas concentration. Fig.1(d) shows the output signal at various CO_2 concentrations. While traditional spectroscopy relies on direct measurement of absorption peaks at specific wavelengths, the OPO sensor output provides multiple information sources across and within a number of detuning peaks containing different spectral information. For instance, in Fig.1(d), the highest output power of the rightmost peak (corresponding to a cavity soliton) decreases as a function of intracavity CO_2 concentrations while other peaks can have the opposite dependence. Cross-referencing other metrics on the peaks can improve the sensitivity and specificity when multiple gases are present. Furthermore, by varying the 2 μm input pump power in addition to the cavity length, we can add another dimension to the output signal for improving sensitivity, as shown in Fig.2. Fig. 2(a) was generated by simulating the output signal of the OPO at two different CO_2 concentrations, 4120 ppm and 412 ppm, then taking their difference. Each "pixel" corresponds to the power difference in the OPO sensor signal at various pump powers (y-axis) and detuning values (x-axis) for these two different concentrations. We also simulated sensing of 392 and 412 ppm CO_2, as well as 412 ppm N_2O to demonstrate multi-species sensing. Fig.2(b) shows the OPO output peaks for various gas concentrations subtracted by a 0 ppm baseline, demonstrating the ability to distinguish between close concentration values and differentiate different gas species. Additionally, due to the nature of half-harmonic generation, the output signal profile is mirrored in input signal depletion, which enables real-time detection of mid-infrared absorptions using a near-infrared detector (Fig.2(c)).

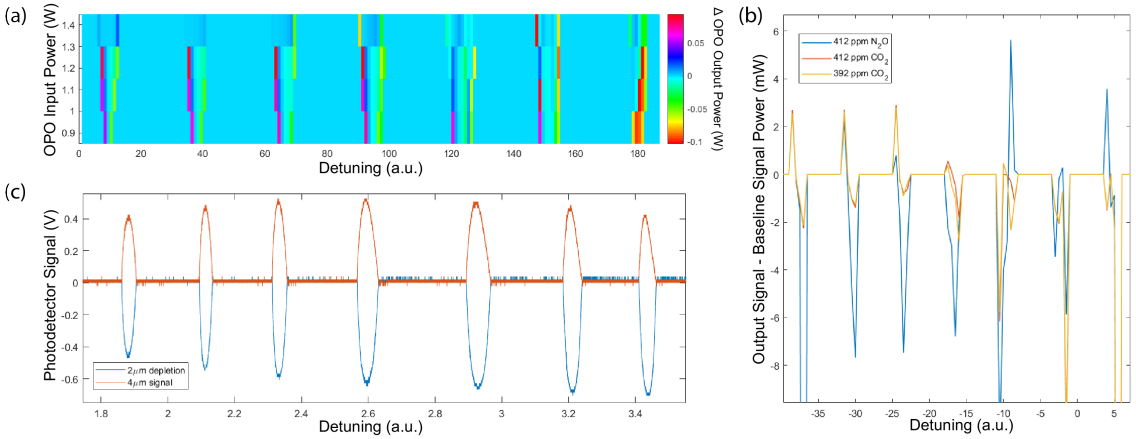

Fig. 2. (a) Difference in simulated OPO output power between sensing 4120 and 412 ppm of CO_2. Each "pixel" corresponds to an output power value difference at different input powers (y-axis) and cavity lengths (x-axis). (b) Simulated OPO output of sensing CO_2 and N_2O at different concentrations at 1.45 W input power. (c) Measured 2 μm depletion signal mirroring 4um output signal.

In summary, by exploiting the nonlinear dynamics of a mid-IR OPO, we introduce and experimentally demonstrate a new technique for multi-species molecular sensing without directly measuring a broadband mid-IR spectrum. The current experimental setup enables broadband mid-infrared gas sensing from ∼3.5 μm to ∼4.8 μm, but this range can be extended by modifying dispersion and loss within the cavity. The OPO sensor offers promising advantages in comparison with direct spectroscopic techniques such as its ability to perform fast, reliable real-time sensing with little disruption to the sensor itself. With significant progress on parametric nanophotonics [12], our sensing scheme can enable compact on-chip multi-species gas sensors without requiring mid-IR lasers and detectors. Our results suggest that the OPO sensor can enable multi-species, broadband, real-time gas sensing for applications such as exhaled breath analysis.

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